(19) JAPANESE PATENT OFFICE (JP)
(12) OFFICIAL GAZETTE FOR UNEXAMINED PATENT APPLICATIONS (A)
(11) Japanese Patent Application Public Disclosure No. 5-139924
(43) Publication Date: June 8, 1993

(51) Int. Cl.[5]	ID Code	Intrabureau Nos.	FI	Technology Indication Area
A 0 1 N 65/00	A	7106-4H		

Request for examination: Not yet submitted
Number of Claimed Inventions: 6

(21) Application No. 3-335687
(22) Filing Date: November 25, 1991

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(54) **[Title of the Invention]**

Plant Blight Damage Control Agent with Natural Ingredients as its Active Ingredients

(57) **[Abstract]**

[Purpose]

[The present invention] relates to a plant blight damage control agent that is applied to plants or soil in order to prevent and control plant blights arising from fungi, bacteria and the like during the cultivation of plants in the fields of agriculture, horticulture and landscape gardening including golf courses and other fields, and provides such an agent that is derived from natural products that have long been consumed as foods by humans and that whose safety has been confirmed.

[Composition]

[The present invention] comprises refined oils extracted from natural products such as cinnamon, cassia, clove, hinoki cypress, eucalyptus, peppermint, spearmint, cumin, star anise, caraway or the like, containing cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole, or has one or more kinds of the aforementioned main components of said refined oils. Alternately, [it] has one or more kinds of ingredients derived from tea such as extracts from tea or tea catechins, tea saponins or other products extracted and separated from tea, or a mixture of said refined oils or their main components and said tea extracts or ingredients derived from tea.

[Claims]

[Claim 1]

A plant blight damage control agent that has as its active ingredients natural ingredients containing extracts from aromatizing natural products or one or more kinds of their main components.

[Claim 2]

A plant blight damage control agent that has as its active ingredients natural ingredients comprising refined oils extracted from natural products such as cinnamon, cassia, clove, hinoki cypress, eucalyptus, peppermint, spearmint, cumin, star anise, caraway or the like, containing cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole, or has one or more kinds of their aforementioned main components.

[Claim 3]

A plant blight damage control agent that has as its active ingredients natural ingredients comprising one or more of the following components extracted from natural products: cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole.

[Claim 4]

A plant blight damage control agent that has as its active ingredients natural ingredients comprising one or more kinds of ingredients derived from tea such as extracts from tea or tea catechins, tea saponins or other tea derivatives extracted and separated from tea.

[Claim 5]

A plant blight damage control agent that has as its active ingredients natural ingredients containing tea saponins that are extracted and separated from tea.

[Claim 6]

A plant blight damage control agent that has as its active ingredients natural ingredients characterized in that they comprise a mixture of the active ingredients listed in claims 1 through 3 and the active ingredients listed in claims 4 and 5.

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

This invention relates to a plant blight damage control agent that is applied in order to prevent and control plant blights arising from fungi, bacteria and the like during the cultivation of plants in the fields of agriculture, horticulture and landscape gardening including golf courses and other fields.

[0002]

[Prior Art Statement]

Conventionally, chemically synthesized organophosphorus or organosulphur compounds or other germicides are used in order to prevent and control plant blights during the cultivation of plants in the fields of agriculture, horticulture and landscape gardening including golf courses and other fields.

[0003]

[Problems Which the Invention is Intended to Solve]

However, many of these germicides with superior antimicrobial activity also are highly toxic, and there is outcry over their damage to the ecosystem of the soil and their adverse effects on humans and fish when present in runoff into streams and rivers or underground water, so demands for highly safe alternatives have grown.

[0004]

Regarding the aforementioned problems, the present inventors focused upon the safety of natural products that have been consumed as foods by humans for many years and that whose safety has been confirmed, and also noted that certain kinds of natural products have in the past been found to have antimicrobial activity with respect to *botulinus bacillus*, *Staphylococcus*, *Vibrio parahaemolyticus* and other bacteria that cause food poisoning, and thus determined to develop and provide a natural product that is safe for humans but is also a plant blight damage control agent

derived from natural products that has preventative activity or control activity with respect to plant blight damage.

[0005]

[Means of Solving the Problems]

In order to solve the aforementioned problems, the present invention provides a plant blight damage control agent that has as its active ingredients natural products that are extracts or components of aromatizing natural products along with extracts or components of tea.

[0006]

To wit, the present invention is a plant blight damage control agent that has as its active ingredients natural ingredients containing one or more kinds of extracts from aromatizing natural products or their main components. As the active ingredients, those that have the most marked plant blight damage control (antimicrobial) activity are those that have the main components of cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole, so having one or more kinds of these main components is extremely effective as a plant blight damage control agent.

[0007]

In addition, cinnamon oil, cassia oil, clove oil, hinoki oil, peppermint oil, eucalyptus oil, spearmint oil, cumin oil, star anise oil, caraway oil and other refined oils extracted from natural products such as cinnamon, cassia, clove, hinoki cypress, eucalyptus, peppermint, spearmint, cumin, star anise, caraway or the like exhibit clear plant blight damage control activity, so having one or more kinds of these refined oils is desirable as a plant blight damage control agent. It is possible to combine one or more kinds of each of the aforementioned main components and the aforementioned refined oils.

[0008]

In addition, as a plant blight damage control agent that has natural ingredients as its active ingredients, ingredients derived from tea such as extracts from tea (including tea seeds; the same hereinafter) or tea catechins, tea saponins or other tea derivatives extracted and separated from tea, particularly tea saponins, exhibit effective activity, so [an agent] having one or more kinds of these can serve as a plant blight damage control agent.

[0009]

By mixing said natural extracts, their main components or refined oils with tea derivatives, tea saponins and other ingredients derived from tea, the plant blight damage control activity can be effectively increased even further.

[0010]

Moreover, to explain the plant blight damage control agent according to the present invention, regarding the plant blight damage control agent according to the present invention, the natural products whose derivatives or components form the active ingredients are those that contain one of the following as components: cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole.

[0011]

Examples of those [natural products] containing cinnamic aldehyde include cinnamon and cassia, those containing eugenol include clove, those containing hinokitiol include hinoki, those containing cineol include eucalyptus, those containing menthol include peppermint, those containing carvone include spearmint, those containing cuminal include cumin and those containing anethole include star anise and anise.

[0012]

Cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole extracted from the dried or fresh leaves, stems, fruit, flowers or other parts of these natural products by means of hot water or ethanol, methanol, acetone, ether or other organic solvents, those that are distilled by water vapor distillation or other methods, or those that are obtained by separation and purification by normal methods from oils obtained by water vapor distillation may be used as the active ingredients of this plant blight damage control agent.

[0013]

Next, the tea used for the extraction or extraction and separation of the active ingredients of the plant blight damage control agent according to the present invention is not particularly limited as long as it is included within the true teas, so green tea or other unfermented teas, oolong tea or other partially fermented teas and black tea or other fermented teas may be used. The extracts from tea are defined to be those that are extracted from these teas by means of hot water or ethanol, methanol, acetone or other hydrophilic organic solvents.

[0014]

The tea catechins referred to here include epigallocatechin, epicatechin gallate, epigallocatechin gallate or crude products containing these catechins, but the theaflavins which are enzyme-oxidized forms of catechin contained in fermented teas, and the enzyme-oxidized forms of catechins contained in partially fermented teas are also included. Tea crude catechins are obtained by extraction from tea leaves using hot water or ethanol, methanol, acetone or other hydrophilic organic solvents, performing a decaffeinating process by means of chloroform and partitioning and concentrating using ethyl acetate. In addition, by further separating this by means of sample-splitting HPLC, purified extracts of the various catechins are obtained.

[0015]

The tea saponins are defined to be crude products containing various saponins of tea leaves and various saponins of tea seeds, but purified tea-leaf saponins are obtained by extracting tea leaves in methanol or water, performing butanol extraction and separating by means of column chromatography, and purified tea-seed saponins are obtained by extracting tea seeds after degreasing, dissolving the extract in ether and hydrochloric acid and precipitating.

[0016]

Note that the methods of extracting and separating tea catechins and tea saponins described above are merely single examples of methods of obtaining the purified extracts, and the methods of extracting and purifying the tea components used as the active ingredients of this plant blight damage control agent are in no way limited to these methods.

[0017]

Examples of the plant pathogens to which the plant blight damage control agent of the present invention may be applied include *Rhizoctonia*, *Pythium*, *Curvularia*, *Helminthosporium*, *Pyricularia* and others, but they are not limited to these. In addition, examples of the plants subject to control of pathogens include grasses, rice and others, but [the agent] is also effective on other fruits and vegetables.

[0018]

The composition of this plant blight damage control agent and the amount applied will vary depending on the pathogen subject to control, the type of plant and the amount of time applied, so they are not particularly limited. In addition, regarding the form of usage, any of the liquid form, gel form, or solid form are possible so there are no particularly limitations, and it can be used when adsorbed to a polymer absorbent or zeolite or other mineral.

[0019]

[Function of the Invention]

By means of the present invention, it has become clear that the following components extracted from natural products: cinnamic aldehyde, eugenol, hinokitiol, cineol, menthol, carvone, cuminal or anethole, or extracts from aromatizing natural products such as refined oils extracted from cinnamon, cassia, clove, hinoki cypress, eucalyptus, peppermint, spearmint, cumin, star anise, caraway or the like have superior antimicrobial activity with respect to plant pathogens, and tea derivatives or tea components also have antimicrobial activity, and moreover mixed compositions of these also have marked antimicrobial activity, yet these active ingredients are components derived from natural products that have long been used as food and drink by humans, so their adverse effects on the environment are few and they are safe for humans, and thus a plant blight damage control agent that is ideally suited as a plant blight damage preventative agent and control agent can be provided.

[0020]

[Preferred Embodiments]

Here follows a detailed explanation of the antimicrobial activity of the plant blight damage control agent of the present invention using preferred embodiments as example, but the present invention is in no way limited to these preferred embodiments as long as they do not diverge in substance.

[0021]

[Preferred Embodiment 1]

We examined the growth inhibiting activity of extracts and components of aromatizing natural products with respect to plant pathogens. We tested the activity of extracts of cinnamon, cassia, clove, eucalyptus, peppermint, spearmint, cumin, star anise, caraway and hinoki along with cinnamic aldehyde, eugenol, menthol and anethole components obtained from cinnamon extracts, clove extracts, peppermint extracts and star anise extracts.

[0022]

As the various extracts of natural products, we used cinnamon oil, cassia oil, clove oil, eucalyptus oil, peppermint oil, spearmint oil, cumin oil, star anise oil, caraway oil and hinoki oil obtained from the dried bark of cinnamon, dried leaves of cassia, dried flower buds of clove, fresh leaves of eucalyptus, dried leaves of peppermint, dried entire plant of spearmint, fruit of cumin, dried fruit of star anise, fruit of caraway, and trunk of hinoki, respectively.

[0023]

The cinnamic aldehyde used was obtained by adding to cinnamon oil two times its weight of acidified sodium sulfite, then adding dilute ethanol and boiling, cooling, filtering, rinsing with 80% ethanol and then adding the same weight of sulfuric acid after drying. The eugenol used was obtained by dissolving clove oil in three times its weight of 10% sodium hydroxide, percolating in ether to remove terpenes and then adding dilute sulfuric acid to the alkaline solution and then performing vacuum distillation. The menthol used was obtained by cooling and centrifuging peppermint oil. The anethole used was obtained by cooling and crystallizing star anise oil.

[0024]

Three plant pathogens were used to test the antimicrobial activity: *Rhizoctonia solani*
[External 1]

kühn

(hereafter called pathogen R), *Pythium aphanidermatum* (hereafter called pathogen P), *Curvularia* spp. (hereafter called pathogen C).

[0025]

The test of the growth inhibiting activity of these extracts and components with respect to lawn pathogens was performed by means of the agar-agar dilution method using a potato sucrose culture medium at three different concentrations (50, 500 and 5000

ppm) for each pathogen, with three runs each. At each concentration, the growth inhibition rate (%) was found from the degree of elongation of the mycelial threads of the pathogens into areas where none was added. An inhibition rate of 100% indicates zero elongation, while an inhibition rate of 50% indicates that the elongation of mycelial threads is 1/2. Note that pathogen P was tested after 24 hours of culture at 30°C, while pathogens R and C were tested after 3 and 6 days of culture, respectively. In addition, the aforementioned extracts and refined oil components were emulsified by means of a sucrose fatty acid ester, diluted to the stipulated concentration and then used in the testing.

[0026]

Tables 1, 2 and 3 show the growth inhibiting activity of the various test materials with respect to pathogens R, P and C. From these results, one can see that they exhibited superior antimicrobial activity in that growth of each of the pathogens was completely inhibited at concentrations of 500 ppm and 5000 ppm.

[0027]

[Table 1]

= Growth inhibition rate of extracts and components of specific natural products with respect to pathogen R =

Test material \ Concentration	50	500	5000
(Extracts)			
· Cinnamon oil	64	100	100
· Cassia oil	51	100	100
· Clove oil	54	100	100
· Hinoki oil	92	100	100
· Peppermint oil	0	55	100
· Eucalyptus oil	0	50	100
· Spearmint oil	0	37	100
· Cumin oil	0	57	100
· Star anise oil	0	75	100
· Caraway oil	0	45	100
(Components)			
· Cinnamic aldehyde	85	100	100
· Eugenol	57	100	100
· Menthol	47	100	100
· Anethole	10	100	100

[0028]

[Table 2]

= Growth inhibition rate of extracts and components of specific natural products with respect to pathogen P =

Test material \ Concentration	50	500	5000
(Extracts)			
· Cinnamon oil	9	100	100
· Cassia oil	23	100	100
· Clove oil	0	100	100
· Hinoki oil	100	100	100
· Peppermint oil	0	60	100
· Eucalyptus oil	0	63	100
· Spearmint oil	17	69	100
· Cumin oil	0	40	100
· Star anise oil	0	43	100
· Caraway oil	0	57	100
(Components)			
· Cinnamic aldehyde	19	100	100
· Eugenol	23	100	100
· Menthol	0	100	100
· Anethole	4	100	100

[0029]

[Table 3]

= Growth inhibition rate of extracts and components of specific natural products with respect to pathogen C =

Test material \ Concentration	50	500	5000
(Extracts)			
· Cinnamon oil	52	77	100
· Cassia oil	0	52	100
· Clove oil	35	79	100
· Hinoki oil	100	100	100
· Peppermint oil	0	51	100
· Eucalyptus oil	0	40	100
· Spearmint oil	0	48	100
· Cumin oil	0	63	100
· Star anise oil	0	38	100
· Caraway oil	0	49	100
(Components)			
· Cinnamic aldehyde	19	100	100
· Eugenol	23	100	100
· Menthol	0	100	100
· Anethole	0	100	100

[0030]

[Preferred Embodiment 2]

We examined the growth inhibiting activity of tea extracts and tea components with respect to plant pathogens. We tested the growth inhibiting activity of extracts of green tea, oolong tea and black tea along with tea crude catechins and tea-seed saponins.

[0022]

The various extracts of green tea, oolong tea and black tea were obtained by extracting leaves of each type of tea for 10 minutes in 20 times their weight of hot water and extracts resulting from vacuum freeze drying were used. The tea crude catechins used were obtained by extracting green tea in 10 times its weight of hot water, and after vacuum concentration, further extracting this in a chromatography column with a solution of

distilled water and 15% acetone, and then performing further concentration and freeze drying. In addition, tea-seed crude saponins are obtained by extracting tea seeds with alcohol after degreasing.

[0032]

The plant pathogens subject to testing were the same three lawn pathogens as in preferred embodiment 1. In addition, testing of the growth inhibiting activity was performed according to the method of preferred embodiment 1.

[0033]

Table 4 shows the growth inhibition rates of the various test materials with respect to pathogen R, Table 5 shows the growth inhibition rates with respect to pathogen P and Table 6 shows the growth inhibition rates with respect to pathogen C. From these results, each of the test materials exhibited growth inhibiting activity with respect to each of the pathogens, and it was found that the growth inhibition rates increased as the concentration of application increased.

[0034]

[Table 4]

= Growth inhibition rate of tea extracts and tea components with respect to pathogen R =

Test material \ Concentration	50	500	5000
· Hot-water extract of green tea	0	3	60
· Hot-water extract of oolong tea	0	31	85
· Hot-water extract of black tea	0	3	81
· Tea crude catechins	0	3	89
· Tea-seed crude saponins	37	67	75

[0035]

[Table 5]

= Growth inhibition rate of tea extracts and tea components with respect to pathogen P =

Test material \ Concentration	50	500	5000
· Hot-water extract of green tea	0	20	88
· Hot-water extract of oolong tea	0	13	100
· Hot-water extract of black tea	0	25	100
· Tea crude catechins	0	19	99
· Tea-seed crude saponins	0	5	65

[0036]

[Table 6]

= Growth inhibition rate of tea extracts and tea components with respect to pathogen C =

Test material \ Concentration	50	500	5000
· Hot-water extract of green tea	0	3	43
· Hot-water extract of oolong tea	0	0	41
· Hot-water extract of black tea	0	5	43
· Tea crude catechins	3	15	72
· Tea-seed crude saponins	19	69	89

[0037]

[Preferred Embodiment 3]

We examined the growth inhibiting activity of mixtures of the aforementioned extracts of stipulated natural products and tea extracts with respect to pathogen R and pathogen P. We prepared 17 different compositions by taking the five components of hot-water extract of oolong tea, cinnamon oil, clove oil, peppermint oil and hinoki oil that were used in preferred embodiments 1 and 2 and varying the content of each (oolong tea extract 20%-50%, cinnamon oil 20%-40%, peppermint oil 10%-40%, hinoki oil 20%-40%). Using these compositions, we examined the growth inhibiting activity with respect to the lawn pathogens of pathogen P and pathogen R used in preferred embodiments 1 and 2. The test of activity was performed by means of the agar-agar dilution method using a potato sucrose

culture medium at three different concentrations with three runs each. At each concentration, the growth inhibition rate for each pathogen was plotted on a logarithmic scale to find the median effective dose ED_{50}, which was compared against the ED_{50} of each refined oil alone to determine whether or not the oolong tea extract has a synergistic effect. Note that the culture conditions for the various pathogens are the same as those of preferred embodiment 1.

[0038]

Table 7 shows those of the 17 combinations that exhibit the synergistic effect of oolong tea with respect to pathogens P or R, along with their activity.

[0039]

[Table 7]

Comparison of the growth inhibiting effects on pathogen R and pathogen R of individual substances and mixed compositions

Test material (composition)	ED_{50} with respect to pathogen R	ED_{50} with respect to pathogen P
· Oolong tea extract 100%	1136	1333
· Cinnamon oil 100%	36	63
· Clove oil 100%	47	108
· Peppermint oil 100%	425	356
· Hinoki oil 100%	26	33
· Mixed composite A	19	55
· Mixed composite B	27	26

[0040]

Mixed composite A listed above, consisting of 20% oolong tea extract, 40% cinnamon oil and 40% clove oil, exhibited the most marked inhibiting effect with respect to pathogen R. Mixed composite B listed above, consisting of 20% oolong tea extract, 10% cinnamon oil, 30% peppermint oil and 40% hinoki oil, exhibited the most marked inhibiting effect with respect to pathogen P.